Synthesis and Characterization of $(La_{1-X}Sr_X)(Ga_{1-Y}Mg_Y)O_3$ Particles by Spray Pyrolysis

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ABSTRACT

Spherical $La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.2}O_{2.8}$ precursor powders were synthesized by ultrasonic spray pyrolysis using aqueous solution of citric acid and of metal nitrate. As-prepared powders obtained by this method have a spherical morphology with a submicron size. The calcination behavior of the precursor powders of the above phases were examined in the temperature range of 1073 –1773 K in an air atmosphere. XRD, SEM, DTA-TG and BET analysis were used for determination of the composition, morphology, particle size and surface area. XRD revealed that as-prepared powders were crystallized to perovskite structure after the sintering at 1573 K for 10 h.

INTRODUCTION

Solid oxide fuel cell (SOFC) as an energy conversion device has attracted much attention because of its higher conversion efficiency and environmental friendship. A typical high-temperature SOFC used 8 mol% yttria- stabilized zirconia (YSZ) with a fluorite structure as an electrolyte [1-3]. A strontium-and-magnesium-doped lanthanum gallate (LSGM) ceramics are known to have superior oxygen ion conducting properties [4-6] as compared to yttria-stabilized zirconia electrolytes. In recent years, several researchers have synthesized LSGM ceramics by mainly using the conventional solid-state reaction, sol-gel [7,8], Pechini method [9]. Ultrasonic spray pyrolysis is well known as an useful tool for the rapid production of multi-component powders in a continuous process. A precursor solution is misted into aerosol droplets that are introduced into a electric furnace where the aerosol droplets undergo drying, droplet shrinkage, solute precipitation, decomposition and sintering to form final particles. The advantage of spray pyrolysis process is as follows; (a) spherical morphology, (b) narrow particle size distribution, (c) easy preparation of the powder with the complex composition, (d) relatively homogeneous composition. In this work, we prepared $La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.2}O_{2.8}$ precursor powders with submicron size, good crystallinity, spherical morphology and non-aggregation by ultrasonic spray pyrolysis. Characteristics of as-prepared particles such as crystallinity, morphology were investigated.

EXPERIMENTAL

A stoichiometric amount of metal citrate were used as starting materials for the synthesis of $La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.2}O_{2.8}$ powders and they were dissolved in distilled water with nitric acid. The concentration of the total metal nitrate was 0.1 M. A spray pyrolysis system was used in this study, consisting of ultrasonic generator, furnace with two different thermal zones and cyclone for produced powders capture. The starting solution was misted at a frequency of 2.4 MHz by an ultrasonic nebulizer (HM-2412, Honda Electronics). The generated mist was carried to an electric furnace by air carrier gas with a rate of 0.7 dm^3/min. The furnace set point temperature for these experiments was varied between 1073 K and 1173 K. Structural properties of the products were investigated by XRD (M3X, MacScience), surface morphology by SEM (S-2300, Hitachi) and specific surface area (SSA) by the BET (Trystar, Shimadzu) method.

RESULTS AND DISCUSSION

Figure 1 shows typical particle morphology of as-prepared powders by SEM photograph and particle size distribution. As-prepared powders have a spherical particle shape with a mean diameter of 1 μm. The particle size ranged 0.5 to 1.8 μm. The value of geometrical standard deviation ($\sigma_{g)}$) calculated from the particle size distribution was 1.36, suggesting that the particle size distribution was narrow. Such morphology is sometimes observed when a ultrasonic spray pyrolysis was employed. SSA was 4.55 m^2/g estimated from BET method. Figure 2 shows particle morphology calcined at various temperatures. After calcinations at 1273 K, spherical particles with diameters of grains ca. 0.8 μm were bonded to one another. After calcinations at 1673 K, flake-like

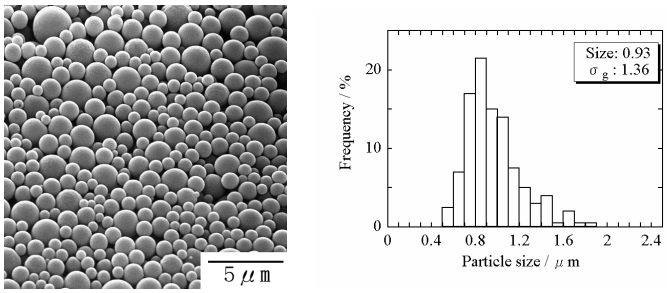

Fig.1 SEM photographs and particle size distribution of As-prepared powders.

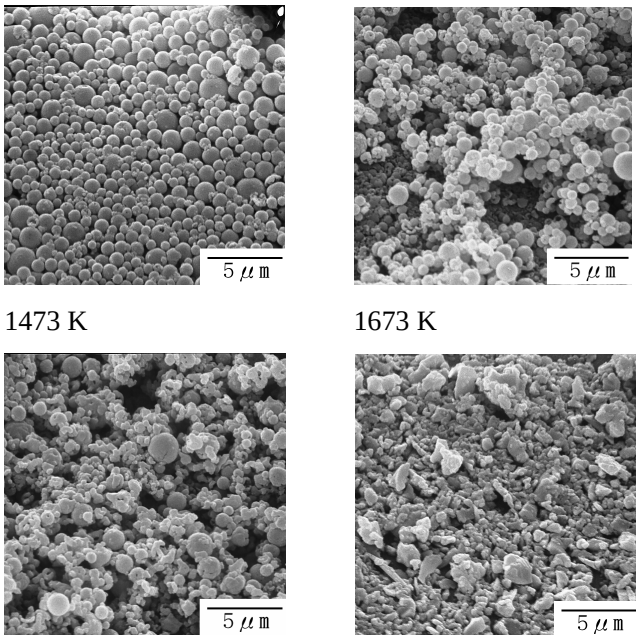

Fig.2 Particle morphology calcined at various temperatures.

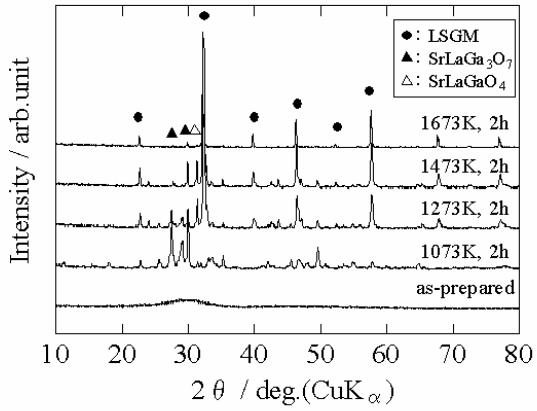

Fig.3 XRD patterns of LSGM powders calcined at various temperatures.

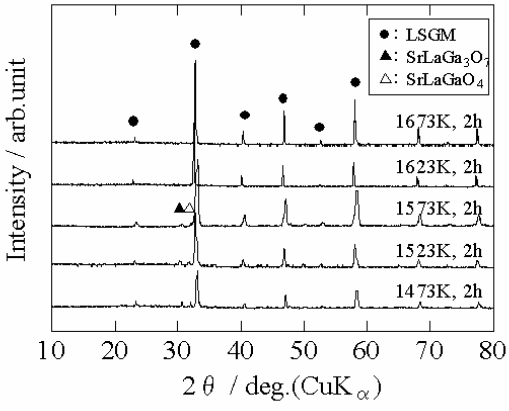

Fig.4 XRD patterns of LSGM pellets sinterd at various temperatures.

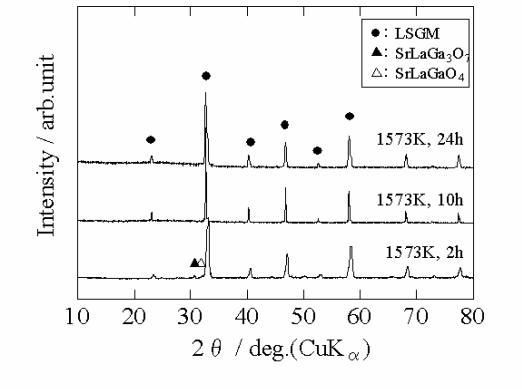

Fig.5 XRD patterns of LSGM pellets sinterd for various times.

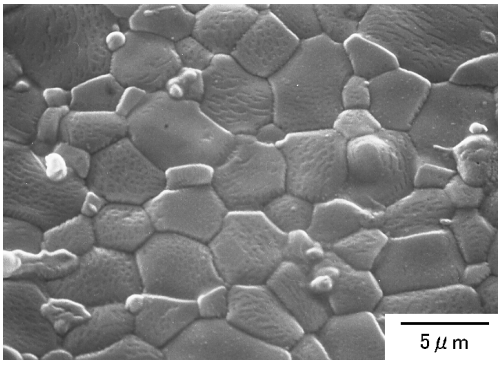

Fig.6 SEM photographs of LSGM sinterd pellet obtained at 1573 K for 2 h.

particles with size of 1-3 μm were formed.

Figure 3 shows X-ray diffraction patterns of as-prepared powders and samples calcined for 2 h at different temperatures. It can be seen from the XRD patterns, the perovskite phase has existed in the calcined powders, but the impurity phases exist clearly as well. Note that the XRD patterns of the samples calcined at 1073 K presents many intermediate phases. Even after calcinations at 1673 K, the powder was not single phase. In fact, the most intense diffraction peaks of $SrLaGaO_4$ and $SrLaGa_3O_7$ [10] were still detectable in the XRD patterns. Figure 4 shows XRD patterns of pellets sintered for 2 h at different temperatures. The samples can be indexed based on a perovskite type structures after sintering at 1623 K. No impurity-related peaks are observed from the XRD patterns. Figure 5 shows XRD patterns of pellets sintered at 1573 K for different time. The crystal phase of sitered pellet was not single phase for 2 h. In contrast, for more 10 h, the single phase of LSGM was obtained. In order to obtain the single phase of LSGM, it is necessary to sintered for 2 h at a temperature of more than 1623 K, or sintered at 1573 K for time of more than 10 h. Figure 6 shows

SEM photographs of LSGM pellet sintered at 1573 K for 2 h. A sintered pellet which is fine-grained, and has a uniform microstructure was obtained.

CONCLUSION

$La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.2}O_{2.8}$ precursor powders were prepared by the ultrasonic spray pyrolysis. As-prepared particles were spherical which have a diameter of about 1 μm with narrow size distribution. Pure powders could not be obtained even after 2 h calcinations at 1673 K. In fact $SrLaGaO_4$ and $SrLaGa_3O_7$ phases were detected by XRD patterns in all calcined powders. However, XRD revealed that as-prepared powders were crystallized to perovskite structure after the sintering at 1623 K for 2 h or 1573 K for 10 h. A sintered pellet which is fine-grained, and has a uniform microstructure was obtained.

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